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Effect of Simulated Acid Rain and Heavy Application of Fertilizers on Two Andisols and Their Effect on Crop Growth

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Summary

The effects of simulated acid rain and the heavy application of fertilizers on acidification of allophanic Zao Andisol and non-allophanic Kawatabi Andisol were investigated in two studies. One was carried out using a 21 cm height and 16.5 cm diameter column, and a rain simulator machine. The other, was done using a stirred-flow reaction system to determine the SO_4^{2-} adsorption capacities of Andisols. For both allophanic and non-allophanic Andisols, the amount of soil extractable SO_4^{2-} increased with depth, but the amount of SO_4^{2-} extracted in Zao soil was much higher than that in Kawatabi soil. The rate of acidification of Kawatabi soil due to acid input was higher than that for Zao soil. The amount of M KCl extractable aluminium was higher than that of water extractable Al, and increased when soil pH decreased. With fertilizer treatments of Kawatabi soil, we found a significant ($P<0.05$) reduction of root growth of barley. Amounts of SO_4^{2-} adsorbed in Zao soil were much higher than those in Kawatabi soil, which was more sensitive to acid inputs. The amount of M KCl extractable Al in Zao soil was lower than that in Kawatabi soil ; however, in both soils it increased with a decrease of soil pH. It was suggested that if soil pH of Zao soil decreased to low values, the amount of M KCl extractable aluminium could be reached to toxic levels for crop growth.

Key words : Andisol, crop growth, heavy fertilization, simulated acid rain

Anthropogenic and volcanic emissions of SO_2 in Asia for 1987-1988 were estimated on a $1^0 \times 1^0$ grid. Anthropogenic sources were estimated to be 31.6 Tg of SO_2 with the regions' volcanoes emitting an additional 3.8 Tg (Ardnt et al., 1997). As a consequence, the atmosphere has been polluted and strong acid deposition has been fallen in many parts of Asia. Soil acidification due to acid deposition has been shown to be a major environmental problem in many areas of

Asia, and has been the focus of many reports (Katsuno et al., 1995; Liao Bohan et al., 1998; Larssen et al., 2000). Additionally, the heavy application of fertilizers has accelerated soil acidification in many agricultural areas where the soil has the possibility of becoming acid with high aluminium concentrations. In Japan, the acidification of both allophanic and non-allophanic Andisols has been accelerated by the recent increased input of acidulent ions through acid precipitation and/or heavy application of acid-forming fertilizers (Matsuyama et al., 1999). Such soils have become strongly acidic with high amounts of M KCl extractable Al, with aluminium toxicity of crops being a severe occurrence.

Soils have a buffering capacity that mediates the impact of acid deposition, but this capacity can become exhausted leading to soil acidification. According to Yoshida and Kawahata (1988), soil acidification due to acid deposition is considered to be accompanied by increased leaching of base cations such as Ca, Mg and K from the soil rhizosphere.

Soils often have ability to adsorb SO_4^{2-} from acid precipitation and this process appears an important part of the buffering power against the impact of acid deposition. Attempts to classify soils in terms of their susceptibility to acidification by acid inputs have included estimates of their SO_4^{2-} adsorption capacity (Harrison et al., 1989). The various mechanisms responsible for sulfate retention by soils are: 1) electrostatic attraction by positively charged sites (Rajan, 1979), 2) specific adsorption onto Fe and Al surface materials involving ligand displacement (Rajan, 1979), 3) immobilization by soil organisms through the conversion of inorganic sulfate to organic sulfate compounds (Swank et al., 1984), and 4) the precipitation of basic aluminium sulfate minerals (Nordstrom, 1982).

Sulfate dynamics of soils can be described either by precipitation/dissolution of Al-hydroxysulphate (Prenzel, 1994; Alewell et al., 1995) or by sorption isotherms (Johnson et al., 1993; Wesselink et al., 1994; Alewell et al., 1995). Attempts to classify soils in terms of their susceptibility to acidification by acidic inputs have included estimates of their SO_4^{2-} adsorption capacity (Harrison et al., 1989). According to Cronan et al. (1977), in soils that have low capacity to adsorb sulfate, the input of SO_4^{2-} from the atmosphere results in accelerated cation leaching; whereas, adsorption of SO_4^{2-} in soils prevents leaching (Singh et al., 1980).

Thus, the objective of this work aimed to evaluate the effect of simulated acid rain and the heavy application of fertilizers on the acidification of allophanic and non-allophanic Andisols and the resulting effects on crop growth.

Materials and methods

I. Column experiment

A column experiment was carried out in a green house at the Experimental Farm, Graduate School of Agricultural Science, Tohoku University, Oosaki, Japan. Non-allophanic Andisol (Kawatabi soil), and allophanic Andisol (Zao soil) were used. Some chemical properties of the soils used are shown in Table 1. Kawatabi soil had more organic matter and exchangeable Ca content than did Zao soil.

PVC pipes of 21 cm height and 16.5 cm diameter with their bottom sealed with a permeable sheet were used as the columns. Three kilograms of air-dried soil (25% soil moisture) was packed in each column.

Treatment simulated two rain types and the heavy application or no soil application of fertilizers. Deionized water without soil fertilization was used as the control treatment. Acid rain was prepared by diluting an M H_2SO_4 solution and adjusting the final pH to 4.0. This solution contained approximately 55.1 mM l^{-1} of SO_4^{2-}. Amounts of 7.2, 3.6 and 3.6 g kg^{-1} air dried soil of N, P_2O_5, and K_2O were applied in the treatments that received fertilizers. Treatments were symbolized as: control treatment (C), acid rain treatment (AR), control treatment with fertilization (C+F), and acid rain treatment with fertilization (AR+F). Deionized water and acid solution were sprayed over the columns using a rain simulation machine (DIK-6000-S) (Plate 1), which was set to simulate a rain of 40 mm.day^{-1}, with a droplet size of about 1.7 mm diameter. Solutions were sprayed overnight for 12 hrs with an interval of 3 days between each spraying period. Volumes of leachate solution from columns were measured after each spray and the electrical conductivity (EC), pH, and Ca, Mg and K concentrations were measured.

C and N contents of the soil samples were measured with a Sumika NC-80S analyzer, and the organic matter content was calculated by multiplying the amount of organic carbon (%) by 1.724. Soil exchangeable Ca, Mg and K were extracted with 0.05 M ammonium acetate: 0.0114 M $SrCl_2$ and analyzed by atomic absorption spectroscopy (AAS) (Hitachi Z-6100). 0.01 M NaOH extracta-

Table 1. Chemical Properties of Used Soils

Soil	pH H_2O	OM %	EC dS/m	Exchangeable cations (cmol kg^{-1})[(a)]			
				Ca	Mg	K	Na
Kawatabi soil	6.01	16.2	0.034	10.0	0.2	0.9	1.8
Zao soil	5.99	4.0	0.072	3.8	0.6	1.3	1.7

[(a)] Ca, Mg, K and Na were determined with 0.05 M NH_4Ac

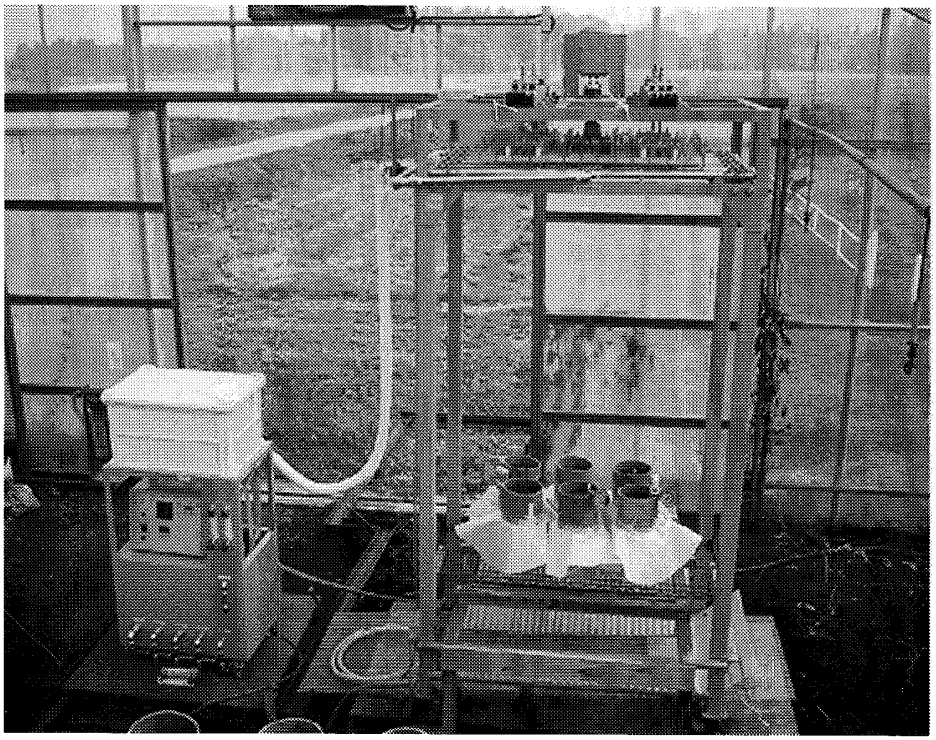

PLATE 1. View of the column in the rain simulator machine (DIK-6000-S)

ble soil SO_4^{2-} was determined according to Fox et al. (1964) and soil exchangeable Al was extracted by M KCl (McLean, 1965) and by water using a soil : solution ratio of 1 : 10. Concentrations of Aluminium in M KCl extracts were analyzed by AAS, while that in water extracts was analyzed by a pyrocatechol violet method (McAvoy et al., 1992). Soil pH was measured using a soil : water ratio of 1 : 2.5.

After spray treatments, soil was divided into different depths; and to evaluate the acidic condition of each depth, barley seedlings were grown for 7 days. Root lengths of barley were measured after harvest.

Sulfate adsorption experiment

The second experiment was carried out to evaluate the differences in the SO_4^{2-} adsorption capacity of the Andisols used in the column experiment.

A stirred-flow reaction cell was used in this experiment, and was similar in design to the reaction chamber used by Dahlgren and Saigusa (1994). Three solutions were used : one was the same acid solution as for the column experiment. The other two were a diluted solution of hydrochloride acid (pH 4), and deionized water used to determine the amount of SO_4^{2-} released from soils. The procedure was as follows : all solutions were pumped into the chamber containing two grams of soil at a constant flow rate of 1.5 ml min^{-1}. The volume of the solution in the chamber was 40 ml. The contents of the chamber were continuously stirred with a magnetic stir star, resulting in a suspension. Soil was retained in the chamber

with a 47 mm diam., 0.45 μm filter, and effluent solution was collected with a fraction collector that was set to collect 10 ml of solution per tube. SO_4^{2-} concentrations of effluent solution were measured by ion chromatography.

This experiment was carried out for 800 minutes, and the amounts of SO_4^{2-} adsorbed in the soils were calculated from the difference between the SO_4^{2-} concentration of the influent solution and the SO_4^{2-} concentration of the effluent solution minus the amount of SO_4^{2-} released from soils in hydrochloride acid solution or deionized water.

Results and discussion

Column experiment

The chemical characteristics of the leachates from Kawatabi and Zao soils are shown in Table 2. During the experimental period, the total volume of each solution sprayed was of 780 mm, and the total volumes of solution leached from the soils were of 497 and 550 mm for Kawatabi and Zao soils, respectively. Thus they were 64% and 70% of total volume of each solution, respectively, sprayed. The total input of SO_4^{-2} to each of the soils during the experimental period was of 250 mg kg^{-1} soil. For both soils, no significant differences were found between C and AR treatments for leachate EC. The mean values of EC in these treatments were approximately of 0.4 and 0.3 dS m^{-1} for Kawatabi and Zao soils, respectively, and were of 17.0 and 14.9 dS m^{-1} in the C+F and AR+F treatments, respectively, of Kawatabi soil. On the other hand, those from Zao soil were of 19.9 and 18.8 dS m^{-1}, respectively.

Table 2. Chemical Chraracteristics of Leachates of Kawatabi and Zao Soils

Treatment/ soil	Total leachated ml	EC (dS m^{-1})			pH			meq l^{-1}			
		min	Max	mean	min	Max	mean	Ca^{2+}	Mg^{2+}	K^+	SUM[a]
Kawatabi soil											
C	497.0	0.1	0.8	0.4	6.09	7.12	6.58	21.1	7.0	0.4	28.5
AR		0.1	0.8	0.4	6.17	7.06	6.63	22.9	7.6	0.5	31.0
C+F		2.0	45.8	17.0	4.32	7.05	5.74	87.9	57.3	34.0	179.2
AR+F		1.5	45.2	14.9	4.54	7.08	5.88	103.5	75.1	34.9	213.5
Zao soil											
C	550.0	0.2	0.6	0.3	6.26	6.99	6.65	5.5	12.0	4.4	21.9
AR		0.2	0.7	0.3	6.14	6.94	6.67	6.8	14.4	5.1	26.3
C+F		2.2	49.5	19.9	4.93	7.19	6.05	54.7	80.7	45.7	181.0
AR+F		1.9	60.1	18.8	4.44	7.19	6.01	51.2	88.6	49.0	188.8

[a] $Ca^{2+}+Mg^{2+}+K^+$

Leachates from Kawatabi soil were more acidic than that from Zao soils. The mean pHs of leachates from Kawatabi soil were of 6.58, 6.63, 5.74 and 5.88 for C, AR, C+F and AR+F treatments, respectively, and those from Zao soil were of 6.65, 6.67, 6.05 and 6.01.

Kawatabi soil lost more Ca than did Zao soil. On the other hand, Zao soil lost more Mg and K than did Kawatabi soil. The total amounts of Ca in leachates from Kawatabi soil were of 21.1, 22.9, 87.9 and 103.5 meq l^{-1} for C, AR, C+F and AR+F treatments, respectively, and those from Zao soil were of 5.5, 6.8, 54.7 and 51.2 meq l^{-1}. The total amounts of Mg in leachates from Kawatabi soil were of 7.0, 7.6, 57.3 and 75.1 meq l^{-1} for C, AR, C+F and AR+F treatments, respectively, and those from Zao soil were of 12.0, 14.4, 80.7 and 88.6 meq l^{-1}. The total amounts of K in leachates from Kawatabi soil were of 0.4, 0.5, 34,0 and 34.9 meq l^{-1} for C, AR, C+F and AR+F treatments, respectively, and those from Zao soil were of 4.4, 5.1, 45.7 and 49.0 meq l^{-1}.

The total amounts of cations, on an equivalent basis, in leachates from Kawatabi soil were higher than those from Zao soil, and for both soils, the simulated acid rain treatments increased the total amounts of the cation losses. The total amounts of cations in leachates from Kawatabi soil were of 28.5, 31.0, 179.2 and 213.5 meq l^{-1} for C, AR, C+F and AR+F treatments, respectively. On the other hand, those from Zao soil were of 21.9, 26.3, 181.0 and 188.8 meq l^{-1}.

Table 3 presents the effects of simulated acid rain and the heavy fertilization on the amount of exchangeable base elements (Ca, Mg, K, Na). For both soils, the same decrease in the amounts of exchangeable Mg, K and Na were found in the non fertilized treatments (C and AR). However, the amounts of exchangeable Ca in Kawatabi soil decreased much more than that in Zao soil. The decreases of exchangeable Ca of Kawatabi soil were of 1.44 and 1.56 cmol kg^{-1} for C and AR treatments, respectively, and those for Zao soil were of 0.5 and 0.64 cmol kg^{-1}. No decrease in the amount of exchangeable cations was found in treatments that received a heavy application of fertilizers.

The mean values of the total base elements in the original Kawatabi and Zao soils were of 12.9 and 7.3 cmol kg^{-1}, respectively. For Kawatabi soil, these values decreased to 9.4 and 9.3 cmol kg^{-1} in C and AR treatments, respectively. For Zao soil, they decreased to 5.3 and 4.9 cmol kg^{-1}. On the other hand, in treatments that received fertilizers, the total amounts of exchangeable base elements in both soils after the experiment were higher than that before, except for Zao soil in superficial layers (0-1 cm).

The effect of simulated acid rain and fertilization on the amount of SO_4^{2-} extracted in Zao and Kawatabi soils is shown in Fig. 1. In all treatments, the amount of soil extractable SO_4^{2-} increased with depth in both soils ; however, the amount of SO_4^{2-} extracted in Zao soil was much higher than that in Kawatabi soil. The highest amounts of SO_4^{2-} were found at 15-20 cm soil depth under

Table 3. Effect of Simulated Acid Rain and Heavy Fertilization on Amounts of Soil Exchangeable Ca, Mg, K and Na

Soil/Depth	Treatments																			
	C				AR				CF				ARF				C	AR	C+F	AR+F
	Ca	Mg	K	Na	Ca	Mg	K	Na	Ca	Mg	K	Na	Ca	Mg	K	Na	SUM[a]			
cm	cmol kg^{-1}																			
Kawatabi soil																				
Initial (0–20)	10.0	0.2	0.9	1.8	—	—	—	—	—	—	—	—	—	—	—	—	12.9	12.9	12.9	12.9
0–1	8.3	0.2	0.2	0.5	8.1	0.2	0.2	0.4	11.3	0.3	4.1	0.8	9.9	0.2	3.5	1.0	9.1	8.9	16.5	14.5
1–5	8.4	0.2	0.2	0.4	8.3	0.2	0.3	0.4	8.9	0.2	4.3	1.0	8.5	0.2	3.6	0.8	9.3	9.2	14.4	13.2
5–10	8.4	0.2	0.2	0.5	8.4	0.2	0.2	0.4	8.6	0.2	4.9	0.9	8.8	0.2	3.9	0.6	9.3	9.2	14.5	13.6
10–15	8.7	0.2	0.3	0.3	8.6	0.2	0.2	0.5	11.6	0.3	4.9	1.3	8.9	0.2	4.0	0.9	9.5	9.5	18.0	13.9
15–20	9.0	0.2	0.5	0.4	8.8	0.2	0.2	0.5	11.4	0.2	5.1	1.1	9.6	0.2	4.2	0.7	10.0	9.7	17.9	14.7
Zao soil																				
Initial (0–20)	3.8	0.6	1.3	1.7	—	—	—	—	—	—	—	—	—	—	—	—	7.3	7.3	7.3	7.3
0–1	2.9	0.5	0.5	0.4	2.6	0.4	0.3	0.4	4.0	0.2	2.3	0.9	3.6	0.2	2.0	0.7	4.2	3.6	7.4	6.5
1–5	3.2	0.5	0.4	0.4	3.1	0.5	0.4	0.7	4.2	0.2	2.8	0.5	3.6	0.2	2.2	0.6	4.4	4.6	7.7	6.5
5–10	3.3	0.5	0.5	0.5	3.2	0.5	0.3	0.8	4.2	0.2	2.6	0.4	3.6	0.2	2.6	0.4	4.7	4.9	7.5	6.9
10–15	3.4	0.5	0.6	0.3	3.4	0.6	0.3	0.7	4.7	0.2	3.5	0.5	4.2	0.2	3.4	0.7	4.9	5.0	9.0	8.5
15–20	3.6	0.6	0.6	0.5	3.4	0.6	0.2	0.8	5.2	0.2	3.7	0.4	4.7	0.2	3.6	0.5	5.3	4.9	9.6	9.0

[a] Ca+Mg+K+Na

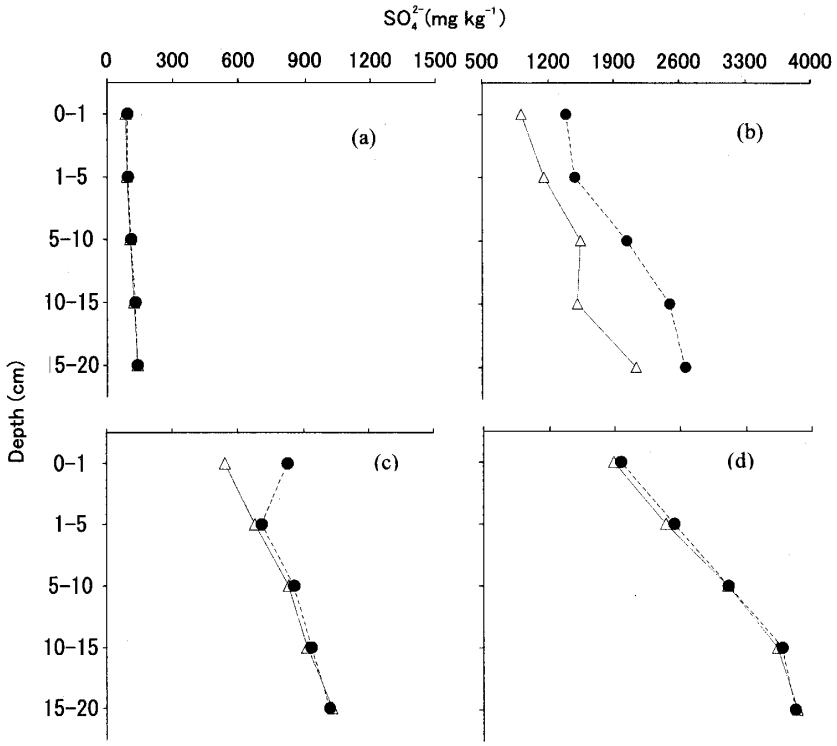

FIG. 1. Effect of simulated acid rain and heavy fertilization on 0.01 M NaOH extractable SO_4^{2-} of (a) Kawatabi soil unfertilized treatment, (b) Kawatabi soil fertilized treatment, (c) Zao soil infertilized treatment, (d) Zao soil fertilized treatment.
(△) control; (●) acid rain

AR+F treatment for both soils, and were of 2,020 and 3,840 mg kg^{-1} in Kawatabi and Zao soils, respectively. For Zao soil, no differences were found in amount of SO_4^{2-} extracted between C and AR treatments, except for in the superficial layer (0–1 cm) of unfertilized treatment. On the other hand, for Kawatabi soil, the amount of SO_4^{2-} extracted in AR treatment was higher than that in C treatment, except for at 15–20 cm soil depth.

In both soils, the heavy fertilization reduced soil pH much more than the simulated acid rain (Fig. 2). The averages of soil pH were reduced 0.16 and 0.22 units in C and AR treatments of Zao soil, and 0.8 and 0.9 units in C+F and AR+F treatments, respectively. For Kawatabi soil, the average soil pH was reduced 0.51 and 0.56 units in C and AR treatments, and 1.35 and 1.43 units in C+F and AR+F treatments, respectively. For both soils, the effect of simulated acid rain was more remarkable at 0–1 cm depth, in which soil pH was reduced 0.34 and 0.67 units in Zao and Kawatabi soils, respectively.

Despite that allophanic Andisols are considered to have little amount of KCl extractable Al, in the soils that received fertilizer, the amount of aluminium extracted by M KCl solution was much higher than that extracted by water, and increased with a decrease of soil pH (Figs. 2 and 3). The amount of aluminium

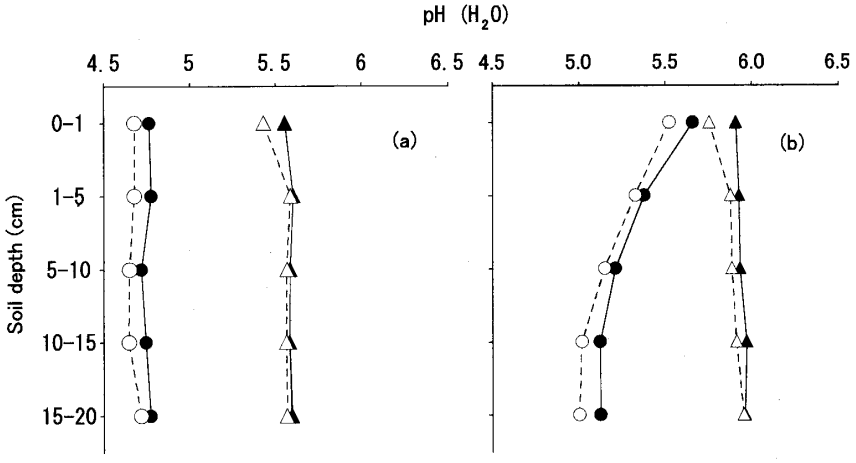

FIG. 2. Effect of simulated acid rain and heavy fertilization onsoil pH of (a) Kawatabi soil and (b) Zao soil. (▲) C, (△) AR, (●) C+F and (○) AR+F treatments

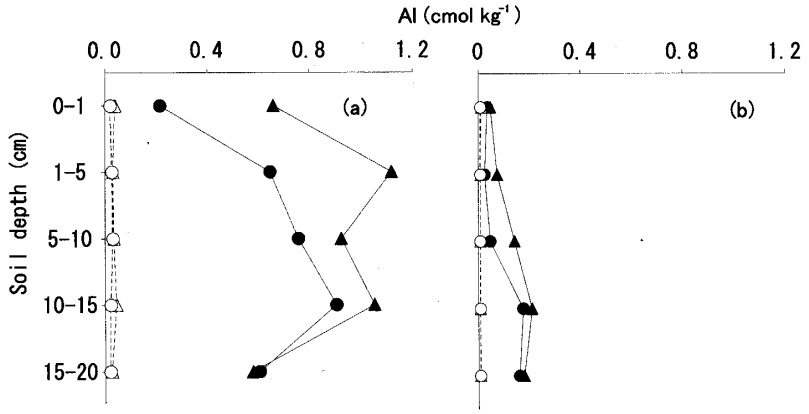

FIG. 3. Effect of simulated acid rain on and M KCl extractable Al of (a) Kawatabi and (b) Zao soils. (○) and (●) are H_2O and M KCl extractable Al of C + F treatment, and (▲) and (△) are the H_2O and M KCl extractable Al of AR+F treatment.

extracted in Zao soil by water extraction was almost 0.01 cmol kg^{-1} in AR+F and C+F treatments, respectively, and the amount extracted by M KCl ranged between 0.03 and 0.21 cmol kg^{-1} in the same treatments. Application of fertilizer reduced the pH of Kawatabi soil more than of Zao soil; consequently, the amount of M KCl extractable Al in Zao soil was lower than that in Kawatabi soil. Despite this, the amount of M KCl extractable Al in Zao soil increased with the decrease of soil pH. However, the origin of M KCl extractable Al in allophanic Andisols is unclear and requires further study. In this study, when soil pH ranged from 5.5 to 5.0 the amount of M KCl extractable Al in Zao soil considerably increased, and it was suggested that at lower pHs the amount of M KCl extractable Al could be increased to levels toxic for crop growth.

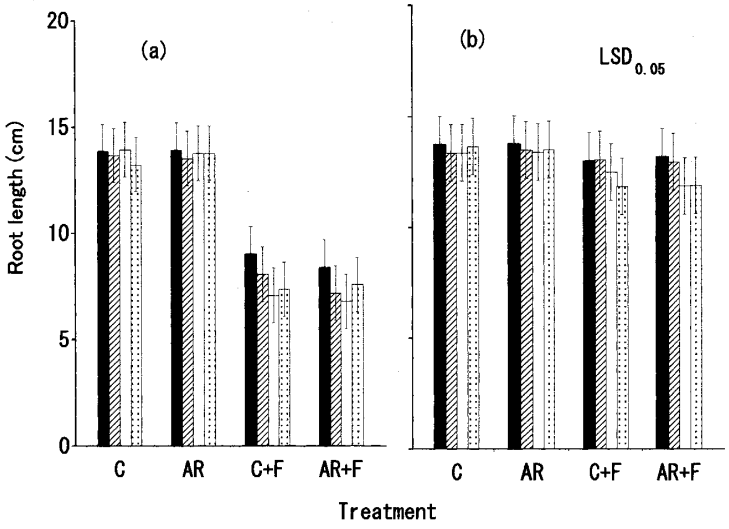

FIG. 4. Effect of simulated acid rain and heavy fertilization on root length of barley grown in soil samples collected from (■) 1-5 cm, (▨) 5-10 cm, (□) 10-15 cm and (⊡) 15-20 cm depth of (a) Kawatabi and (b) Zao soils.

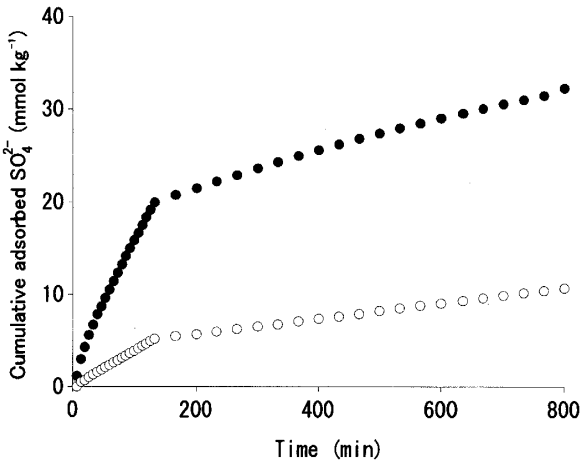

FIG 5. Commulative amounts of SO_4^{2-} adsorbed by (○) Kawatabi Soil and (●) Zao soil.

No significant ($p<0.05$) differences were found between C and AR treatments for root growth of barley, which was significantly ($p<0.05$) reduced in Kawatabi soil acidified by heavy application of fertilizers (Fig. 4), though no significant ($p<0.05$) reduction was found in Zao soil acidified by heavy fertilization.

Sulfate adsorption experiment

During the first minutes, the release of SO_4^{2-} from both soils was higher than the input leading to values of Sin/Sout lower than 1. However, this value increased to above 1 with time due to the increased adsorption of SO_4^{2-} in the

soils. As shown in Fig. 5, different patterns of SO_4^{2-} retention were found for Zao and Kawatabi soils. The amounts of SO_4^{2-} adsorbed in Zao soil were higher than those adsorbed in Kawatabi soil. The total amount of SO_4^{2-} adsorbed in Zao soil during the 800 minute experimental period was of 31.2 mmol kg^{-1}, while Kawatabi soil adsorbed only 10.3 mmol kg-1 for the same period. This explained the high buffering capacity of Zao soil observed in the column experiment. Higher losses of SO_4^{2-} from Kawatabi soil, mainly in fertilized treatments, seemed to be responsible for the lower exchangeable base element content and high acidification found in this soil.

Conclusions

Kawatabi soil was more acidified by simulated acid rain and heavy fertilization than Zao soil.

Kawatabi soil had a lower sulfate adsorption capacity than did Zao soil and was more sensitive to acid inputs through experimental simulated acid rain and the heavy application of fertilizers. Probably, the sulfate adsorption/desorption process had an important direct effect on soil acidification.

Allophanic soils are considered to have low acidity and low amounts of M KCl exchangeable aluminium. In this experimental investigation the amount of M KCl extractable Al from Zao soil was lower than that of Kawatabi soil, and increased with the decrease of soil pH. It was suggested that if the soil pH of Zao soil decreased to low values, a high amount of M KCl extractable aluminium can be expected.

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